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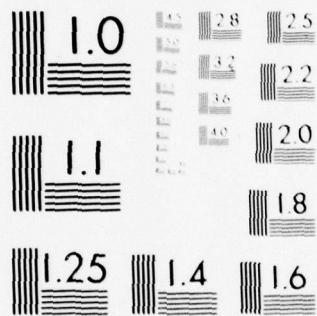
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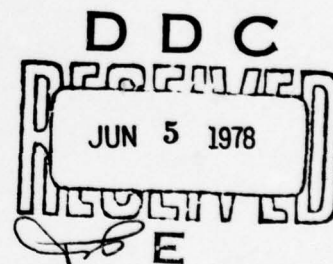
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GAIN MEASUREMENTS OF THE SEQUENCE BANDS
IN A TEA-CO₂ AMPLIFIER

P. Lavigne
J.-L. Lachambre
G. Otis



Centre de Recherches pour la Défense
Defence Research Establishment
Valcartier, Québec

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6 GAIN MEASUREMENTS ON THE SEQUENCE BANDS
IN A TEA-CO AMPLIFIER, (sub 2)

by

10 P./Lavigne, J.-L./Lachambre ■ G./Otis

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RESUME

Après avoir obtenu des coefficients de gain de l'ordre de $1.5\% \text{ cm}^{-1}$ sur les bandes séquentielles $00^0 2-(10^0 1, 02^0 1)_{I, II}$ dans un amplificateur CO_2 TEA, nous avons constaté que le rapport entre le coefficient de gain des bandes séquentielles et celui des bandes régulières ne dépendait pas de la proportion de CO_2 et de He dans le mélange de gaz. Ce rapport s'accroît plutôt avec la quantité d'énergie électrique déposée dans le milieu actif pour atteindre 1/3 à un niveau d'excitation de 140 J/l . Le coefficient de gain relativement élevé des bandes séquentielles laisse entrevoir la possibilité de faire fonctionner un laser CO_2 TEA à des fréquences autres que celles ordinairement émises par les lasers CO_2 TEA et qui permettent une meilleure transmission dans l'atmosphère. Nous avons aussi constaté que la valeur anormalement élevée du coefficient de gain des premières raies de la branche R de la transition régulière à $9.4 \mu\text{m}$ dans les amplificateurs CO_2 TEA était due à l'influence des bandes séquentielles. (NC)

ABSTRACT

After having measured small-signal gains as high as $1.5\% \text{ cm}^{-1}$ in a TEA CO_2 amplifier on the $00^0 2-(10^0 1, 02^0 1)_{I, II}$ sequence bands, we have concluded that the ratio of the gain coefficient of the sequence bands over that of the regular bands was independent of either the CO_2 or He proportion in the gas mixture. This ratio rather increases with the electrical energy dumped into the active medium to reach a value of 1/3 at an excitation density of 140 J/l . Due to the relatively high gain of the sequence bands, it should be possible to have standard TEA CO_2 lasers emit at frequencies where the atmospheric transmission at sea level is higher than at the regular emission frequencies. Furthermore, gain on the sequence bands was responsible for the anomalously high-gain coefficient measured at low J values of the R branch of the regular $9.4\text{-}\mu\text{m}$ transition in TEA amplifiers. (U)

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FIGURES 1 to 6

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1.0 INTRODUCTION

Since the resonance absorption by atmospheric carbon dioxide limits the TEA CO₂ laser radiation propagating along a horizontal path and in clear atmosphere at sea level (Refs 1, 2), the use of the less abundant C¹³O₂¹⁶ and C¹²O₂¹⁸ isotopic molecules has been suggested to achieve better laser transmission in the 8-13 μm atmospheric window (Ref. 3). However, the relatively high cost of these rare CO₂ isotopes prevents their extensive use in high-consumption flowing-gas systems.

Recently, many new emission lines have been observed in CW-CO₂ lasers (Refs 4, 5). These lines which were identified as rotation-vibration transitions in the 00⁰2 - (10⁰1, 02⁰1)_{I,II} bands of the C¹²O₂¹⁶ molecule are referred to as the sequence bands. They emit at frequencies located between ≈ 0.1 and ≈ 20 GHz about the regular CO₂ emission lines. As the energy levels interacting in these newly observed lasing transitions lie about 2400 cm⁻¹ above those of the regular lines, the corresponding concentration of CO₂ absorbers is about 10⁻⁵ less. Laser radiation from these lines would propagate in the atmosphere with absorption losses comparable to those associated with the rare CO₂ isotope laser lines. If one takes into account the influence of the wings of the regular transition, the absorption coefficient of atmospheric CO₂ at the sequence band frequencies should be about an order of magnitude smaller than at the regular frequencies.

The purpose of this document is to examine the possibility of having laser emission at the sequence band frequencies in a TEA-CO₂ amplifier. The gain characteristics of a TEA-CO₂ module are studied as a function of the gas composition, the excitation level and the emission line. The results are compared with the gain characteristics of the regular lines. This work was performed at DREV during the first half of 1977 under PCN 33H01 (formerly PCN 34A01), "Laser Applications in Surveillance Remote Sensing Technology".

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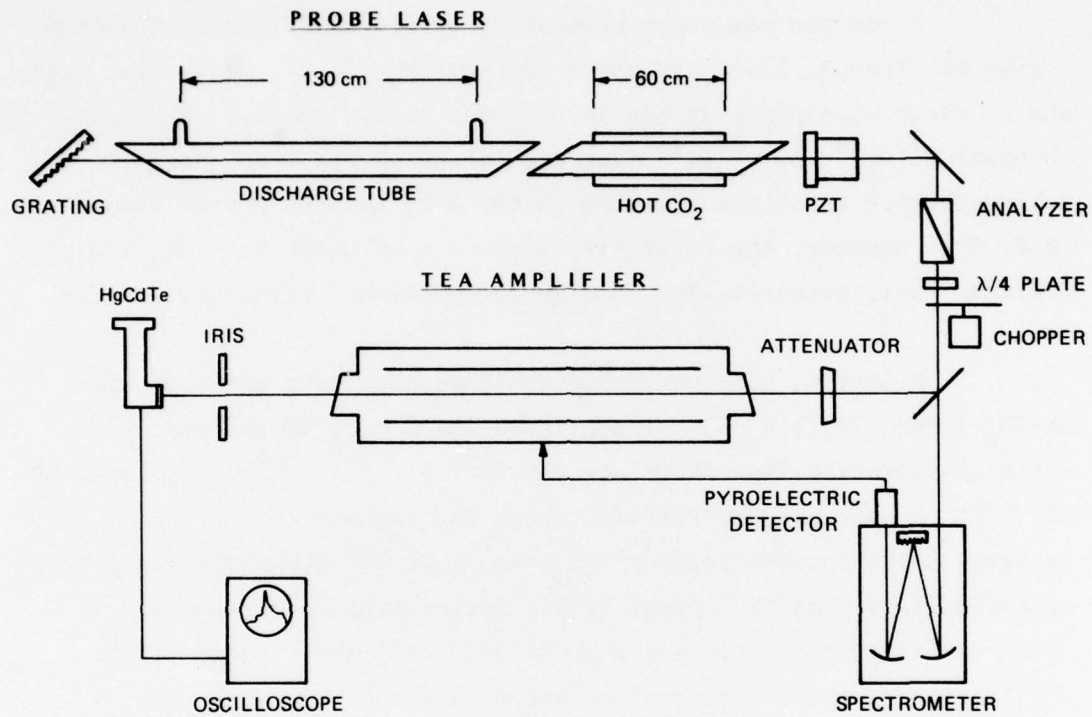


FIGURE 1 - Schema of the apparatus used to measure the gain of the sequence bands in a TEA amplifier module.

2.0 EXPERIMENTAL

Figure 1 illustrates the experimental apparatus. The gain measurements are performed in a $3.8 \times 5 \times 100 \text{ cm}^3$ Laflamme-type TEA module (Ref. 6). A passively tuned discharge circuit, similar to the driving circuit described in Ref. 7, reliably delivers up to 150 J/ ℓ of electrical energy into the active volume. Two slightly tilted NaCl windows seal the Plexiglas box that houses the electrode structure. The gas composition is determined by measuring the flow rate of each constituent of the He-CO₂-N₂ gas mixture.

The low-level signal required for gain measurements is derived from a conventional low-pressure laser. The water-cooled 12-mm I.D. tube has an active length of 130 cm. A 60-cm long cell filled with 40 torr of hot CO₂ at 600 K is inserted into the cavity to allow operation on the sequence bands (Ref. 5). This probe laser can also be operated on the regular bands if we evacuate the absorption cell. The flowing He-CO₂-N₂:80-7-13 mixture is excited with a 16-mA current. The optical cavity is formed by a ZnSe coupling mirror (95% reflectivity) and a 75 ℓ /mm concave grating ($R = 5 \text{ m}$) for line tunability. This laser is stabilized in frequency at the transition center by means of standard cavity dithering techniques. An analyzer followed by a $\lambda/4$ plate optically isolates the source from the amplifier. Part of the signal is directed into a Spex Czerny-Turner spectrometer for frequency monitoring while the remaining beam is further attenuated by a factor of 10 and passed through the TEA amplifier. The amplified signal is limited by an iris for monitoring the central part of the active volume and is detected with a HgCdTe photodiode connected to a Tektronix 7904 oscilloscope through a fast amplifier.

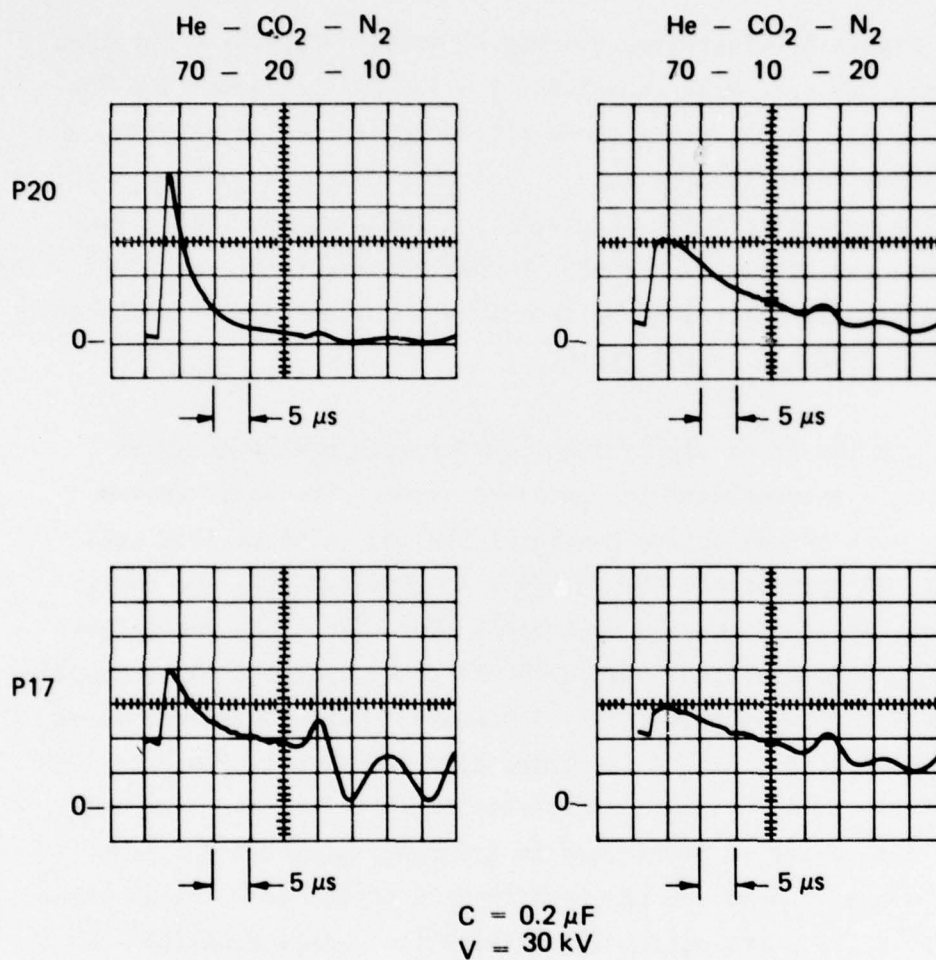


FIGURE 2 - Temporal behavior of the small-signal gain of the regular P20 and of the sequence P17 for two different gas mixtures.

3.0 GAIN MEASUREMENTS

3.1 Time evolution of the gain

Figure 2 illustrates the time evolution of the gain on both the P20 line of the regular bands and the P17 line of the sequence bands of the 10.4- μm transition for two different mixtures at an excitation level of 50 J/l. The gain coefficient is seen to evolve similarly on both transitions. It begins to increase after the same delay, reaches its maximum value at the same time and decays with approximately the same time constant. The fast oscillations on the gain curve, occurring about 20 μs after the firing of the discharge, originate from shock wave perturbations in the probed region (Ref. 8). In N_2 rich mixtures, the pumping effect of excited nitrogen flattens the top portion of the gain curve in a similar way for both transitions. Close examination of the time evolution of the gain coefficients shows that the 00^0_1 and 00^0_2 levels of CO_2 decay towards their equilibrium value with the same characteristic time. This close similarity between the time history of gain coefficients follows from the fast exchange time between the vibrational levels of the antisymmetric stretching mode.

3.2 Influence of the gas composition

The peak-gain coefficient on both the P17 line of the sequence bands and the P20 line of the regular bands is measured with different gas mixtures flowing in the TEA module. With the He proportion fixed at 70%, the gain on both bands increases with the $[\text{CO}_2] / [\text{CO}_2] + [\text{N}_2]$ ratio up to 0.6 and saturates at higher CO_2 proportion as illustrated in Fig. 3a. At an excitation energy density of 50 J/l, the ratio $\alpha(\text{P17}) / \alpha(\text{P20})$ of the gain coefficients stays constant at about 0.17 over the entire studied range.

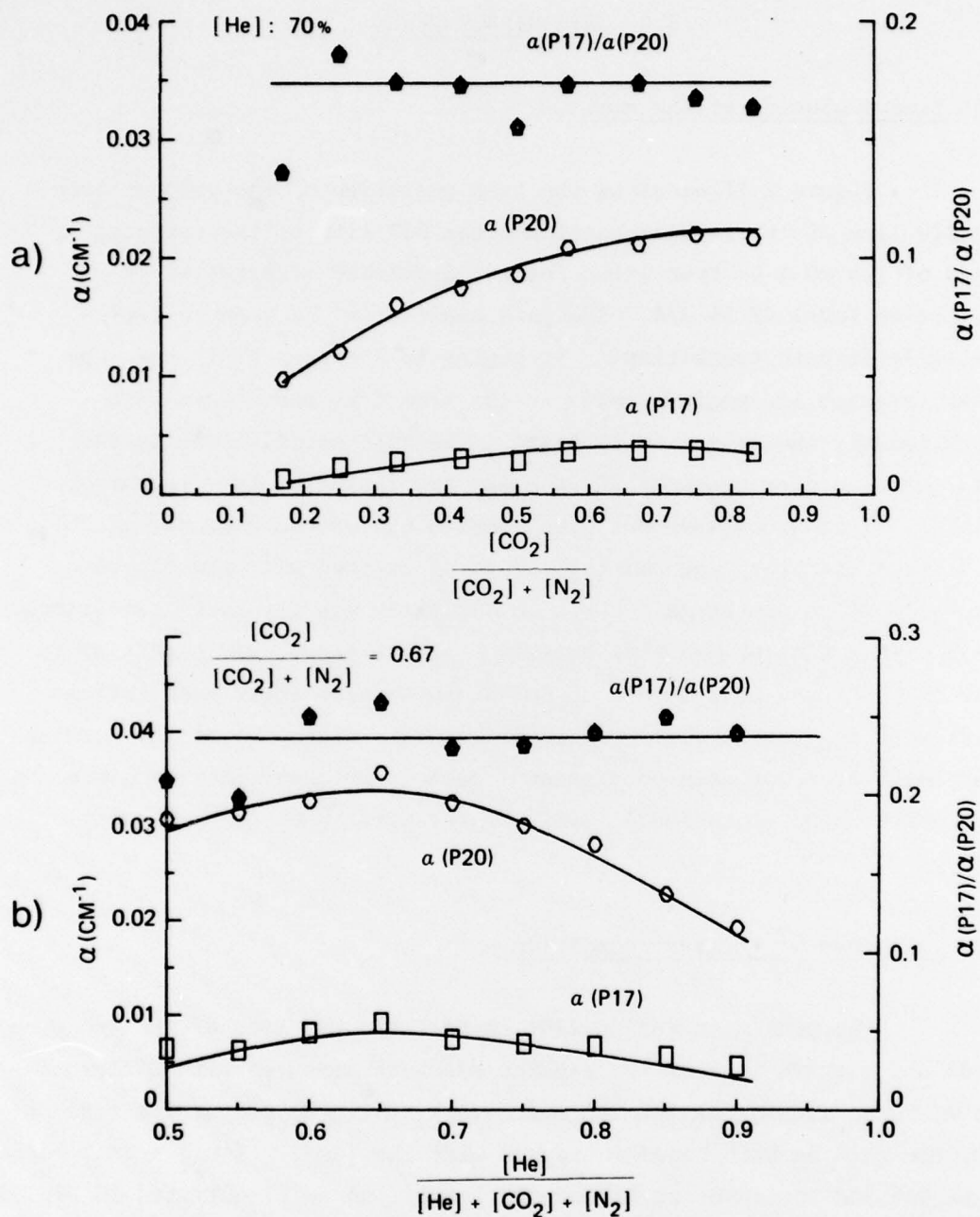


FIGURE 3 - Gain as a function of: (a) the $[\text{CO}_2]/[\text{CO}_2] + [\text{N}_2]$ ratio at an excitation energy density of 50 J/l, and (b) the $[\text{He}]/[\text{He}] + [\text{CO}_2] + [\text{N}_2]$ ratio at an excitation energy density of 85 J/l.

When the $[\text{CO}_2] / [\text{CO}_2] + [\text{N}_2]$ ratio is kept at 67%, both gain coefficients vary similarly with the He content (Fig. 3b). These coefficients increase slightly to reach a maximum with approximately 65% of He proportion and diminish continuously at higher concentrations. In the covered range of He proportions, the $\alpha(\text{P17}) / \alpha(\text{P20})$ ratio stays constant at a value of 0.24 for 85 J/ℓ of electrical energy deposited into the discharge. This indicates that the vibrational temperature of the ν_3 mode is not significantly influenced by the He concentration. The variation of the individual gain coefficient with the He content could be explained by a competitive effect existing between a diminution of the CO_2 concentration and an evolution of the discharge conditions towards a better matching of the driving circuit leading to a more efficient pumping of the active medium.

Contrary to observations made in a low-pressure continuous-wave laser (Ref. 5), the vibrational temperature is not severely affected by the CO_2 content of the active mixture so that optimum gain on the sequence band transitions occurs with the same gas composition as on the regular band transitions. For the type of double discharge used in the experiment, this optimum mixture is typically 70-20-10 (Ref. 8).

3.3 Influence of the electrical excitation

The variation of the gain coefficients with the amount of electrical energy dumped into the discharge has been measured in a He- CO_2 - N_2 gas mixture of 70-20-10. In this experiment, the P20 and P17 lines of the 10.4- μm transitions have been chosen as representative of the regular and sequence-band behavior. As illustrated in Fig. 4, the gain coefficients of both the P17 line (of the sequence bands) and the P20 line (of the regular bands) increase with the pumping energy. The ratio $\alpha(\text{P17}) / \alpha(\text{P20})$ grows monotonically from 0.12 to 0.35 as the excitation energy density is raised from 50 to 140 J/ℓ, if one assumes full transfer into the active volume. With the present

system, it was not possible to further increase the excitation. For a fixed electrical energy, the driving circuit parameters do not seem to affect strongly the $N(00^0_2)/N(00^0_1)$ population ratio of the 00^0_2 and 00^0_1 vibrational levels of CO_2 . This is illustrated in Fig. 4 where two sets of results were obtained by charging different capacitor banks.

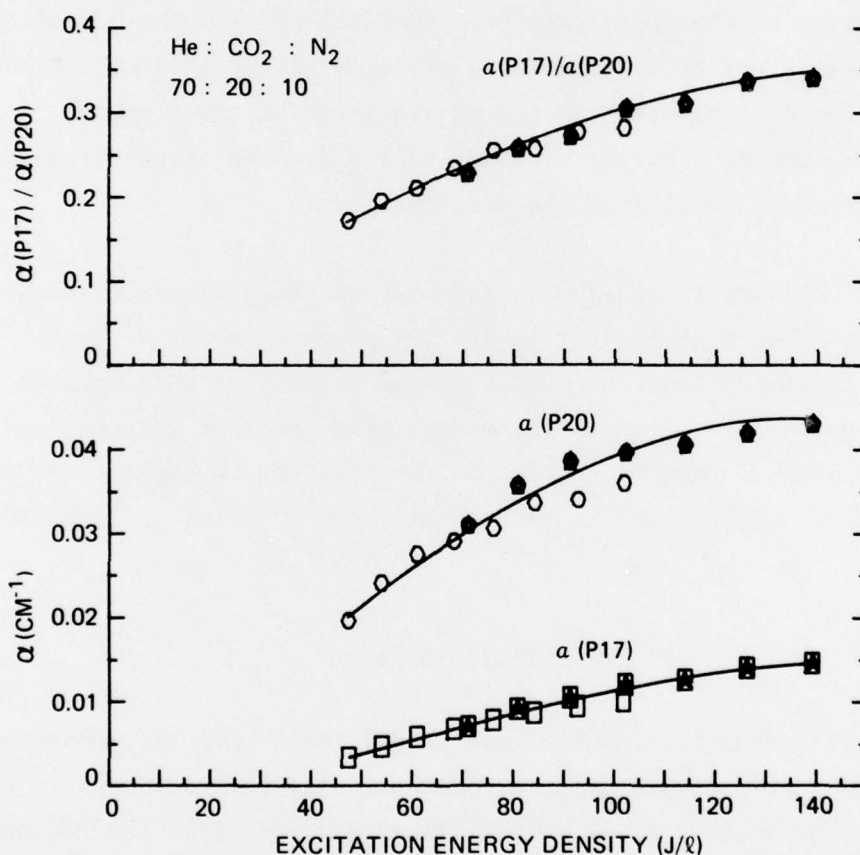


FIGURE 4 - Gain coefficients of the P17 and P20 lines and gain coefficient ratio as a function of the excitation energy density in a 70-20-10: He-CO₂-N₂ gas mixture. The light circles and boxes represent data obtained with a 0.2- μF capacitor bank; the dark ones, data obtained with a 0.3 μF capacitor bank.

The above results can be interpreted in terms of vibrational temperature. If one assumes that a fast equilibrium is set among the antisymmetric stretching levels of the CO_2 molecule and a negligible population of the lower lasing level, the vibrational temperature T_3 of the ν_3 mode can be calculated from the ratio of the population of the 00^02 level over the population of the 00^01 level as deduced from the ratio of the small-signal gain of the sequence bands over the small-signal gain of the regular bands. This temperature is given by:

$$T_3 = \frac{h\nu'_3}{k \ln (\alpha_s \sigma_r / \alpha_r \sigma_s)} \quad (1)$$

where h is the Planck constant; k , the Boltzmann factor; $h\nu'_3$, the energy difference between the 00^02 and the 00^01 levels; α_s and α_r , the small-signal gain coefficients of the sequence and regular bands; and σ_s and σ_r , the radiative cross sections of the sequence and regular bands. The circles in Fig. 5 give the variation of T_3 with the excitation energy as computed from Eq. 1 in the harmonic oscillator approximation, where the ratio σ_s/σ_r amounts to 2.

Alternatively, a value of the vibrational temperature T_4 of the ν_3 mode can be calculated from the ratio of the population of the 00^01 level over the population of the ground state as deduced from the small-signal gain of the regular band and the value of the radiative cross section. This temperature is given by:

$$T_4 = \frac{h\nu_3^0}{k \ln (\alpha_r Q_v / \sigma_r N_0)} \quad (2)$$

where $h\nu_3^0$ is the energy of the 00^01 level and N_0 , the density of CO_2 molecules. Q_v represents the vibrational partition function as calculated from the expression:

$$Q_v = (1 - e^{-h\nu_1/kT_1})^{-1} (1 - e^{-h\nu_2/kT_2})^{-2} (1 - e^{-h\nu_3^0/kT_4})^{-1} \quad (3)$$

where $h\nu_{1,2}$ and $T_{1,2}$ are respectively the energy and the vibrational temperature of the symmetrical stretching and bending vibrational modes of CO_2 . The boxes in Fig. 5 give the variation of T_4 with the excitation energy as obtained by self-consistently solving Eqs 2 and 3 with the value of σ_r given in Ref. 9, and assuming that T_1 , T_2 and the translational temperature are all equal to the rotational temperature that has been experimentally determined around 325 K.

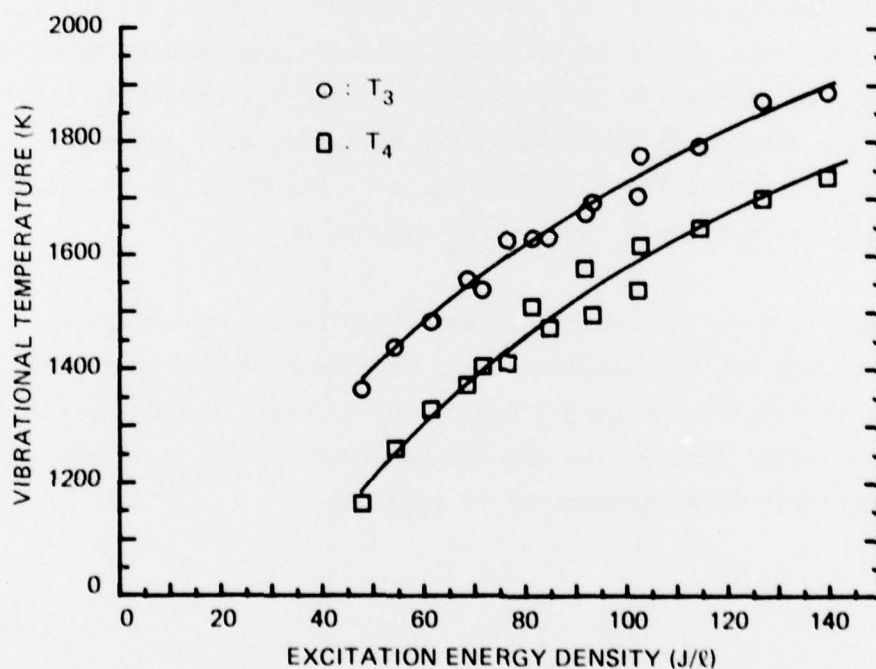


FIGURE 5 - Variation of the temperature of the antisymmetric stretching mode of CO_2 as a function of the excitation-energy density as calculated from the ratio of the gain coefficients (T_3) and from the gain coefficient of the regular band (T_4).

Despite the approximations involved, the agreement between the two curves is quite good indicating that the distribution of the excited molecules in the lowest states of the ν_3 vibrational mode is close to a Boltzmann distribution. Both curves show a similar rate of vibrational temperature increase with the excitation energy. At high excitation levels, the growing rate of T_3 and T_4 diminishes probably following a broadening of the discharge section at high currents (Ref. 8). The 150-K discrepancy between the two curves of Fig. 5 can easily be explained by a 10% error in the experimental results or by an imprecision of 10% in either the value of the radiative cross section of the P20 line or in the value of the ratio σ_s/σ_r .

3.4 J variation of the gain

The absolute value of the small-signal gain coefficient together with its variation with the J number of the different P and R bands allows the determination of the rotational temperature and of the inversion density. Figure 6 shows such a variation measured on both the regular and the sequence bands in an He-CO₂-N₂ mixture of 70-20-10 excited at an electrical energy density of 100 J/l. If one uses a single temperature for the rotational, translational, bending and symmetrical stretching movements, an upper-level population density of $4.4 \times 10^{17} \text{ cm}^{-3}$ and a rotational temperature of 325 K are obtained by curve-fitting the experimental data of the regular transition with the theoretical expression of the gain coefficient (Refs 10, 11). Furthermore, if one assumes that the above temperatures are the same on the sequence bands, a similar gain expression is used to fit the experimental data by varying a factor that takes into account differences in the dipole moment and in the population density. Such fits are represented by the continuous curves in Fig. 6. In all cases, the theoretical description, which is generally satisfactory, shows some particular features that we will now discuss.

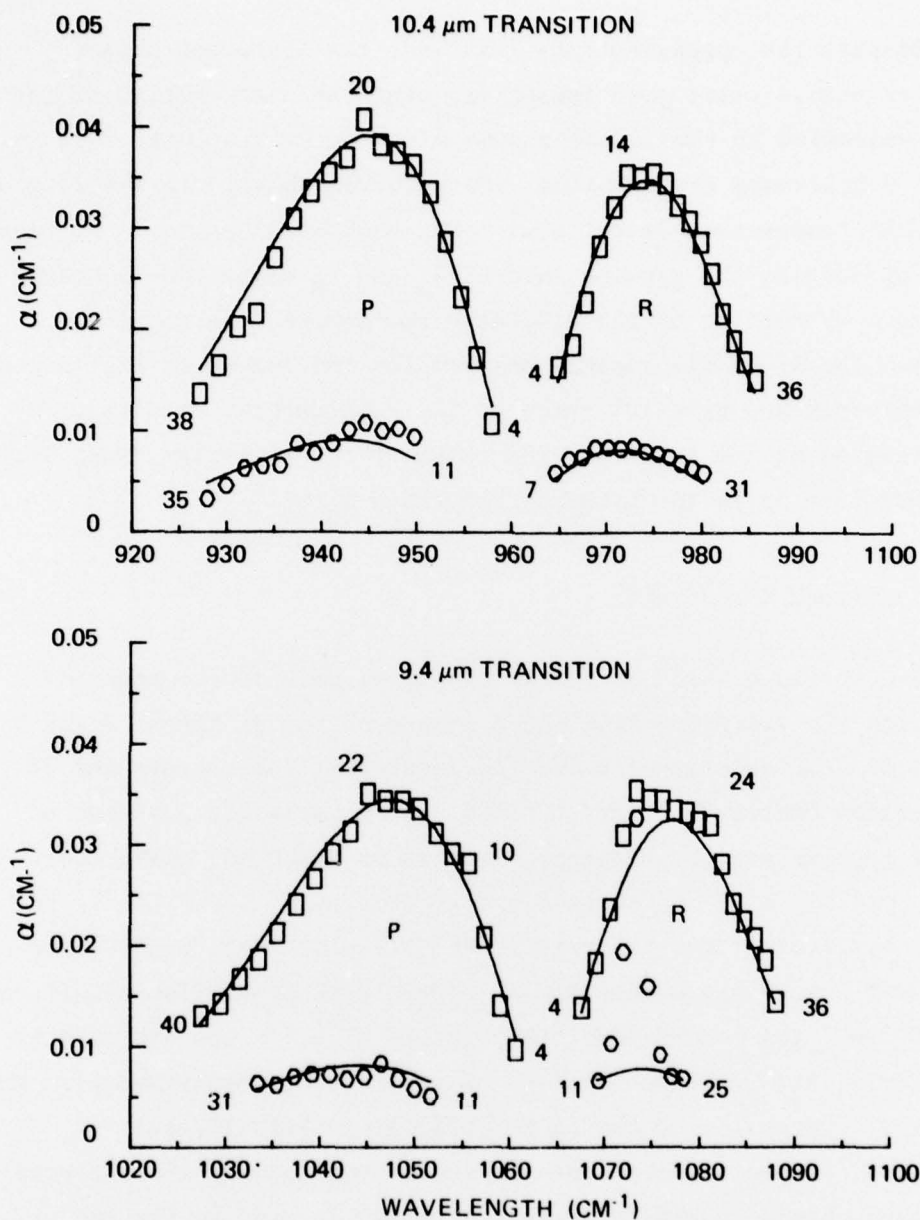


FIGURE 6 - J variation of the small-signal gain in a 70-20-10:He-CO₂-N₂ gas mixture at an excitation energy density of 100 J/l. Boxes represent data on the regular bands and circles, data on the sequence bands. Continuous lines illustrate curve-fitted theoretical expressions.

The regular lines exhibit anomalously high-gain coefficients on the P20 and R14 lines of the 10.4- μm transition and on the P22, P10, R10 to R16 and R24 lines of the 9.4- μm transition. The gain enhancement on the P20 line at 10.4 μm and on the P22, P10 and R24 lines at 9.4 μm has already been reported or predicted by many authors (Refs 12,16). It has been suggested that these lines are overlapped by those of the R band of the $(01^1_1 - 11^1_0)$ transition that have their centre frequencies close to the above-mentioned regular lines. The accuracy of our measurements does not allow us to determine if the gain on the P34 and P28 lines at 10.4 μm is larger than expected, as suggested in Refs 14 and 15, as well as that on the other lines predicted in Ref. 16.

As seen in Fig. 6, anomalously high gain is also observed at a low J value of the R branch of the 9.4- μm transition on both the sequence and regular bands. The correspondence that exists between the two gain curves in that part of the spectrum clearly indicates a gain contribution of both bands at each of the sequence and regular line center frequencies. In fact, wavelength calculations from published vibrational rotational constants (Ref. 17) and measurements (Ref. 18) show that the R13, R15, R17 and R19 sequence lines are respectively 4.5, 2, 0.6 and 3.1 GHz from the R8, R10, R12 and R14 regular lines. Assuming a 4.6-GHz-wide Lorentzian lineshape, it is predicted, as observed, that these lines contribute by 20 to 80% of their gain coefficient value to the gain at the center of the closest neighboring line to the other band. This effect is more apparent on the sequence lines due to the higher gain coefficients of the regular lines. This relative contribution of the sequence lines to the gain of the regular lines increases with the pressure and can explain, as suggested in Ref. 16, the anomalously high gain measured in supra-atmospheric-pressure TEA amplifiers (Ref. 19). At low pressures, where the lines are Doppler broadened and much narrower, this overlapping is much weaker so that it is still possible to selectively filter out the regular lines to get laser action exclusively on the sequence lines in a low-pressure tube.

A contribution from the $00^02 - (10^01 - 02^01)_I$ band has already been suggested to explain the enhancement of the gain on the R14 line at $10.4 \mu\text{m}$ (Ref. 14). Wavelength calculations indicate that the R17 line is the closest one to the sequence band. This transition is about 14.4 GHz off the R14 line, so that it is expected to barely affect its gain. This is confirmed by the absence of a corresponding increase of the measured gain coefficient of the R17 line. This gain enhancement remains difficult to explain as the $(01^11 - 11^10)$ band have no line in this frequency region and the possible $00^03 - (10^02 - 02^02)_I$ contribution should be much weaker than what is observed.

4.0 CONCLUSIONS

In the double-discharge TEA amplifier studied, maximum gain of the $00^02 - (10^01 - 02^01)_{I,II}$ sequence bands of CO_2 has been found to occur in the same gas mixture as the one that optimizes the gain of the $00^01 - (00^01 - 02^00)_{I,II}$ bands. A small-signal gain coefficient as high as $1.5\% \text{ cm}^{-1}$ has been measured on the sequence P17 line. The gain coefficients of the sequence lines have also been observed to increase more rapidly with the excitation energy than those of the regular lines. It is therefore expected that efficient laser emission on the sequence lines would be obtained in TEA amplifiers with high electrical excitation density.

The strong coupling between the 00^01 and the 00^02 levels of CO_2 indicates that the lower members of the chain of the antisymmetric stretching levels form an energy reservoir that feeds the particular state depopulated by stimulated emission. In principle, it should then be possible to extract the same amount of optical energy from any member of the chain. However, particularly in wide-band TEA amplifiers, the insertion losses of the optical filter required to prevent laser emission on the regular lines would probably limit the efficiency of energy extraction on the sequence lines.

5.0 ACKNOWLEDGEMENTS

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"Gain Measurements on the Sequence Bands in a TEA-CO₂ Amplifier"
by P. Lavigne, J.-L. Lachambre and G. Otis

After having measured small-signal gains as high as 1.5 cm^{-1} in a TEA CO₂ amplifier on the $00^0_2-(10^0_1, 02^0_1)_{1,11}$ sequence bands, we have concluded that the ratio of the gain coefficient of the sequence bands over that of the regular bands was independent of either the CO₂ or He proportion in the gas mixture. This ratio rather increases with the electrical energy dumped into the active medium to reach a value of 1/3 at an excitation density of 140 J/L. Due to the relatively high gain of the sequence bands, it should be possible to have standard TEA CO₂ lasers emit at frequencies where the atmospheric transmission at sea level is higher than at the regular emission frequencies. Furthermore, gain on the sequence bands was responsible for the anomalously high-gain coefficient measured at low J values of the R branch of the regular 9.4- μm transition in TEA amplifiers. (U)

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After having measured small-signal gains as high as 1.5 cm^{-1} in a TEA CO₂ amplifier on the $00^0_2-(10^0_1, 02^0_1)_{1,11}$ sequence bands, we have concluded that the ratio of the gain coefficient of the sequence bands over that of the regular bands was independent of either the CO₂ or He proportion in the gas mixture. This ratio rather increases with the electrical energy dumped into the active medium to reach a value of 1/3 at an excitation density of 140 J/L. Due to the relatively high gain of the sequence bands, it should be possible to have standard TEA CO₂ lasers emit at frequencies where the atmospheric transmission at sea level is higher than at the regular emission frequencies. Furthermore, gain on the sequence bands was responsible for the anomalously high-gain coefficient measured at low J values of the R branch of the regular 9.4- μm transition in TEA amplifiers. (U)

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